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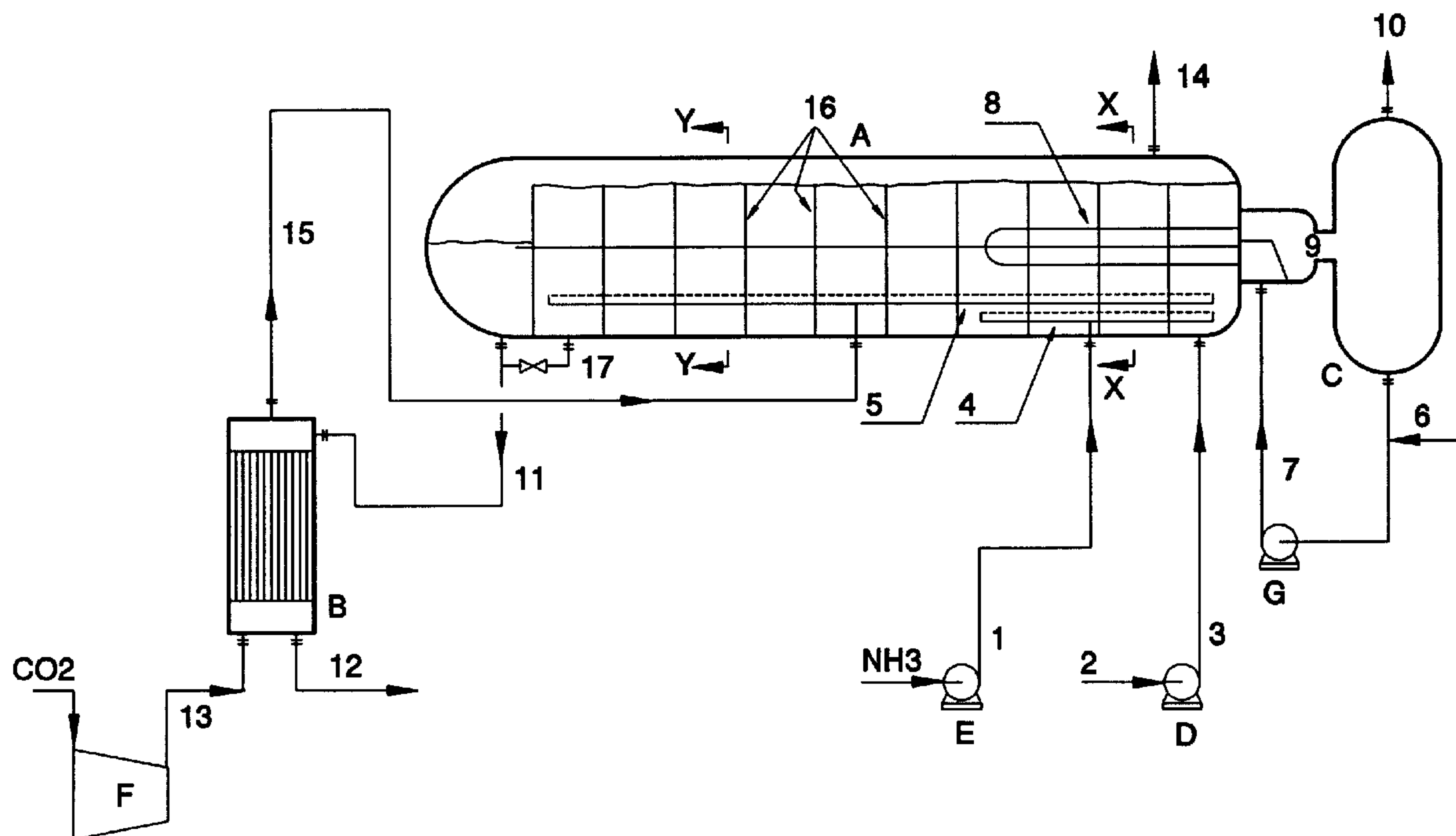
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(54) Titre : METHODE DE FABRICATION D'UREE

(54) Title: METHOD FOR THE PREPARATION OF UREA



(57) Abrégé/Abstract:

The present invention relates to a process and apparatus for the preparation of urea, and in particular for urea obtained from ammonium carbamate formulated from carbon dioxide and ammonia at a pressure of about 125 bar to about 350 bar in a urea synthesis solution in a urea reactor. The reactor defines a horizontally arranged condensation zone and contains a heat exchanger. According to this process, NH_3 and CO_2 are supplied to the reactor and are largely absorbed into the urea synthesis solution. A substantial portion of the heat formed in the condensation is discharged by means of the heat exchanger. The residence time of the urea synthesis solution in the reactor preferably is selected so that at least about 85% of the theoretically obtainable amount of urea is prepared. Thereafter, the urea synthesis solution can be processed into a urea solution or solid urea.

METHOD FOR THE PREPARATION OF UREA

ABSTRACT OF THE DISCLOSURE

5 The present invention relates to a process
and apparatus for the preparation of urea, and in
particular for urea obtained from ammonium
carbamate formulated from carbon dioxide and
ammonia at a pressure of about 125 bar to about
350 bar in a urea synthesis solution in a urea
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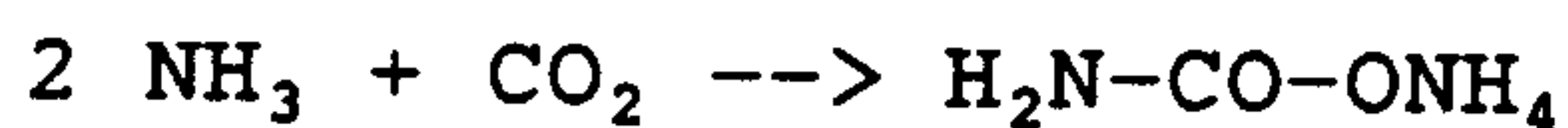
METHOD FOR THE PREPARATION OF UREA

BACKGROUND OF THE INVENTION1. Field of the Invention

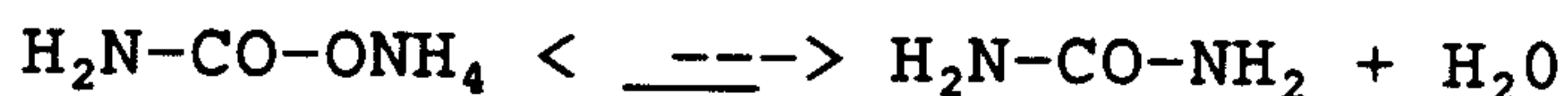
5 The present invention relates to a method for the preparation of urea from ammonia and carbon dioxide.

2. Description of the Related Art

10 When ammonia and carbon dioxide are introduced into a synthesis zone of a urea plant at a suitable pressure (of for example 125-350 atm.) and a suitable temperature (of for example 170-250°C), ammonium carbamate is first formed
15 according to the exothermic reaction:



20 Then urea is formed from the obtained ammonium carbamate through dehydration according to the endothermic equilibrium reaction:



25 The extent to which the conversion to urea takes place is partly dependent on the temperature and pressure employed, and the amount of excess ammonia used. The reaction product obtained is a solution that consists substantially of urea,
30 water, ammonium carbamate and unbound ammonia. The ammonium carbamate and the ammonia are usually removed from the solution and are in most cases returned to the synthesis zone. The synthesis zone may consist of separate zones for the formation of
35 ammonium carbamate and urea. These zones may, however, also be combined in a single apparatus.

A method that is frequently used for the preparation of urea is described in European Chemical News, Urea Supplement of 17 January 1969, pp. 17-20. According to this method, the urea synthesis solution that is formed in the synthesis zone at a high pressure and temperature is transferred to a stripping zone. There, the synthesis solution is subjected to a stripping treatment at a pressure that is substantially identical to that in the synthesis zone by contacting the solution with a countercurrent of gaseous carbon dioxide while supplying heat, so that the majority of the ammonium carbamate present in the solution decomposes to ammonia and carbon dioxide. These decomposition products are stripped from the solution in gaseous form and are evacuated together with a small amount of water vapor and the carbon dioxide used for the stripping. The gas mixture obtained in the stripping treatment is transferred to a condensation zone, in which the majority of the mixture is condensed and absorbed into an aqueous solution produced by the further treatment of the urea-containing solution. Then both the aqueous ammonium carbamate solution formed in this process and the uncondensed gas mixture are passed from the condensation zone to the synthesis zone for the formation of urea. The heat required for the conversion of ammonium carbamate into urea is here obtained through further condensation of the gas mixture, which releases the heat of condensation.

EP-B-155,735 describes a method for the preparation of urea, and reports that a good synthesis efficiency is obtained and the formation of biuret and the hydrolysis of urea in the stripping treatment remain within acceptable limits. Moreover, the gas mixture obtained in the

stripping treatment reportedly is condensed at such a temperature that a considerably smaller heat-exchanging surface area suffices to discharge the heat released, with low-pressure steam of for example 3-5 bar being formed, or steam of a higher pressure, for example of 5-10 bar, being obtained or the heat released being used directly to heat process streams.

According to EP-B-155,735, this is achieved by causing urea to form in the condensation zone as the gas mixture obtained in the stripping operation is condensed. Because of the presence in the condensation zone of relatively large amounts of urea in water, which medium acts as a solvent for the ammonium carbamate formed by the condensation of the gas mixture obtained in the stripping, the heat of condensation and of solution becomes available at a higher temperature level than when this medium is not used. When 30% of the equilibrium amount of urea obtainable has been formed, a heat effect is already clearly noticeable. According to EP-B-155,735 the urea formation is preferably continued to 50-80% of the equilibrium.

According to EP-B-155,735 the condensation zone may be arranged either horizontally or vertically. EP-B-155,735 further mentions that a vertically arranged condensation zone affords the possibility of combining the synthesis zone and the vertical condensation zone in a single apparatus.

JP-A-122,452/84 discloses that a horizontally arranged reactor can be used for urea production. However, JP-A-122,452/84 does not mention combining condensation and synthesis zones. Rather, it describes a method for the preparation of urea, in which a horizontal reactor, and a

separate stripper and condenser are used, the gases from the stripper being condensed in the condenser before the condensate is reintroduced into the reactor.

5 U.S. Patent No. 3,024,280 describes a method for the preparation of urea, in which a long tubular winding reactor having a total length of approximately 140 m is used. CO₂ is added to the urea synthesis solution at various points along
10 the reaction tube while the solution is cooled with the aid of cooling water that passes through the tube jacket.

SUMMARY OF THE INVENTION

15 It is an object of the present invention is to provide an improved method for the preparation of urea.

It is a further object of the present invention to provide a method for the preparation
20 of urea entails lower investment costs.

To accomplish these and other objects, an embodiment of the present invention provides a method for the preparation of urea, the urea being formed along with ammonium carbamate from a
25 reaction between carbon dioxide and ammonia, preferably at a pressure of about 125 bar to about 350 bar in a urea synthesis solution in a urea reactor. The urea reactor preferably contains a horizontally arranged condensation zone which
30 includes a heat exchanger. According to this method, ammonia (NH₃) and carbon dioxide (CO₂) are supplied to the reactor and are largely absorbed into a urea synthesis solution. A substantial portion of heat is generated by condensation.
35 This heat is discharged by, for example, means of the heat exchanger. The residence time of the urea synthesis solution in the reactor is selected

so that at least about 85% of the theoretically obtainable amount of urea is prepared, whereupon the urea synthesis solution can be processed into a urea solution or solid urea.

5 The method according to the present invention is advantageous in that, among other things, the method can be carried out in a plant with substantially lower investment costs, because of the integration of a heat exchanger/condenser in a
10 single reactor. Accordingly, fewer apparatuses and pipelines - which must be resistant to high pressure in a highly corrosive environment - are required. Because the present invention can
15 suitably be (and preferably is) practiced with a condenser/heat exchanger part having a horizontal orientation, a smaller (shorter) plant and plant installations are required. This offers further investment benefits and also promotes safety.

20 Preferably the entire reactor is designed as a horizontally arranged reactor.

 In addition, in accordance with the present invention the supply of the gas phase (CO_2 and NH_3) can then be distributed across the reactor so that a higher degree of conversion can be achieved, if
25 desired. It is also possible to obtain a particular degree of conversion at a lower pressure. In this manner, the operation of a stripper in the process is simplified. Another
30 advantage of a completely horizontally arranged reactor is that it can be started up in a simple manner, because the entire plant can also be operated when the reactor is only partly filled.

 These and other objects, features, and advantages of the present invention will become
35 apparent from the following detailed description when taken in conjunction with the accompanying

drawings which illustrate, by way of example, the principles of the present invention.

BRIEF DESCRIPTION OF THE DRAWING

5 The accompanying drawings illustrate the present invention. In such drawings:

10 FIG. 1 is a schematic view of a high-pressure section of a urea plant suitable for practicing an embodiment of the process of the present invention.

 FIG. 2 is a cross-sectional view (along X-X in FIG. 1) of a reactor suitable for urea plant of FIG. 1 and, in particular, the condensation zone of the reactor.

15 FIG. 3 is a cross-sectional view (along Y-Y in FIG. 1) of the reactor and heat exchanger and, in particular, the condensation zone of the reactor.

20 DESCRIPTION OF THE PREFERRED EMBODIMENTS

 In accordance with an embodiment of the present invention, a method is provided for preparing urea from ammonium carbamate, which is formulated from carbon dioxide and ammonia. This method comprises the steps of supply NH_3 and CO_2 to a synthesis zone of a reactor operated at a pressure of about 125 bar to about 350 bar. The reactor includes a synthesis zone, and a horizontally arranged condensation zone with a heat exchanger disposed therein. Ammonia and carbon dioxide are supplied to the reactor so that the ammonia and the carbon dioxide undergo condensation and are substantially absorbed into a urea synthesis solution, which is maintained in the reactor for a residence time selected so that at least about 85% of a theoretically obtainable amount of urea is prepared. A substantial portion

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of heat formed from the condensation can thereby be discharged to a fluid passing through the heat exchanger.

5 In accordance with a further embodiment, at least a portion of the carbamate is removed from the reactor and decomposed in a stripping zone to form carbamate decomposition products. These products are then passed to the reactor for condensation in the condensation zone, with a
10 substantial portion of heat formed from the condensation being discharged to the fluid passing through the heat exchanger.

The theoretically obtainable amount of urea is determined by the thermodynamic position of the
15 equilibrium and is dependent on for example the NH_3/CO_2 ratio, the $\text{H}_2\text{O}/\text{CO}_2$ ratio and the temperature and can be calculated with the aid of the models as described in for example Bull. of the Chem. Soc. of Japan 1972, Vol. 45, pp.
20 1339-1345, and J. Applied Chem. of the USSR (1981), Vol. 54, pp. 1898-1901.

Preferably the residence time of the urea synthesis solution is selected so that at least
25 about 90% of the theoretically obtainable amount of urea is prepared, and more preferably more than about 95%.

The conversion of carbamate into urea and water in the reactor can be effected by ensuring
30 that the residence time of the reaction mixture in the reactor is sufficiently long. The residence time will generally be more than about 10 min., and preferably more than about 20 min. On the other hand, the residence time also will generally
35 be shorter than about 2 hours, and preferably shorter than about 1 hour. At a higher reactor temperature and pressure, a short residence time

will often suffice to obtain a high degree of conversion.

5 The pressure in the reactor is preferably between about 130 bar and about 210 bar, and the temperature is preferably between about 170°C and about 200°C.

10 The reactor is usually designed to generally have the shape of a wide pipe with a diameter of between about 1 m and about 5 m, preferably between about 2 m and about 4 m. The length of the reactor is generally between about 5 m and about 40 m, preferably between about 10 m and about 25 m.

15 The reactor is generally provided with means that ensure that the liquid flows through the reactor substantially in plug flow. To this end, the reactor is provided with, for example, a structured packing (in one or more places) or with partitions, which divide the reactor into
20 compartments. The compartments are somewhat similar in appearance to "continuously stirred tank reactors" (CSTRs) arranged in series. Although reference is made herein to CSTRs and compartments, these terms are included in the
25 interest of brevity and are not intended to restrict the invention thereto.

The number of compartments or CSTRs in the reactor arranged in series is generally greater than 2 and preferably greater than 5. In general
30 the number of compartments (CSTRs) will be smaller than 40, preferably smaller than 20.

The compartments are preferably defined by substantially vertical partitions. The partitions preferably have a surface area that is at least
35 about 50% of the area of the vertical cross-sectional area of the horizontally placed reactor, and more preferably at least about 85% of

the vertical cross-sectional area of the reactor. Preferably the area of the partitions is at most about 98% of the vertical cross-sectional area of the horizontally placed reactor.

5 Preferably turbulence in the reactor compartments is provided by introducing a gas via a distributing device, for example via a pipe having holes therein, the pipe being disposed at or near the bottom of the reactor. Ammonia, carbon
10 dioxide and/or inert gas can suitable be used as the introduced gas.

 The liquid ammonia preferably is introduced into the reactor in the portion thereof that also contains the both the condensation zone and the
15 heat exchanger. The CO_2 and any gaseous ammonia present preferably are fed into all the compartments of the reactor, but a larger proportion, preferably more than about 60%, is preferably fed to the portion of the reactor that
20 contains both the condensation zone and the heat exchanger. Inert gas can be fed into all or any combination of the compartments. The inert gas will often be present in recycle streams, together with CO_2 and gaseous ammonia.

25 The gas phase that is fed to the reactor can be divided optimally between the series-arranged CSTRs by suitable dimensioning of the distributing device. This makes it possible, for example, to minimize the amount of non-condensable gases in
30 the off-gas of the last CSTRs. In this manner, the vapor pressure of the condensable components is maximized in that part of the reactor where the thermodynamically determined equilibrium is approached. This means that, at a given total
35 pressure (i.e., the sum of the vapor pressure of the condensable components plus the pressure of the non-condensable components), the temperature

in this part of the reaction zone is maximized, as a result of which a higher degree of conversion is obtained. Alternatively, this arrangement is also suitable for minimizing the total pressure at a given temperature, so that for example a better performance of the stripping operation can be realized.

The heat or energy that is discharged in the heat exchanger of the reactor is generally more than about 125 kWh per ton of urea produced. In general, the energy will be less than about 800 kWh per ton of urea produced.

The heat released in the reactor can then be transferred to and discharged by water or other fluid that is passed through tubes of the heat exchanger and is in the process converted into low-pressure steam of preferably about 3 bar to about 10 bar, and more preferably about 4 bar to about 7 bar. The heat can also be discharged by passing a process stream that is to be heated through it, for example a urea solution that is to be evaporated at about 2 bar to about 8 bar or a urea solution that is to be expanded at about 15 bar to about 40 bar. The heat exchanger is preferably disposed in the condenser portion of the reactor. The condenser portion preferably accounts for about 10% to about 70% of the total length of the reactor, and more preferably about 30% to about 50% of the total length.

Preferably the condensation zone and heat exchanger of the reactor are designed as a so-called submerged condenser, in which a portion of the gaseous mixture to be condensed, the ammonia, and dilute carbamate solution are fed into the shell side of a shell-and-tube heat exchanger. The heat of solution and condensation released is transferred to and discharged by a

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medium flowing through the tubes, for example water, which is converted into low-pressure steam.

The method according to the invention can be used in a so-called conventional urea process, but
5 is preferably used in a stripping process as described in ECN Urea Supplement of 17 January 1969, pp. 17-20, in Hydrocarbon Processing of July 1975, pp. 102-104, or in Nitrogen, May-June 1990, pp. 22-29. In this preferred embodiment of the method according to the invention the urea synthesis solution formed in the reactor is fed to a stripper, where carbamate is decomposed, whereupon the gases obtained are returned to the
15 reactor.

The decomposition of the ammonium carbamate present in the urea synthesis solution is generally effected by supplying heat. If only heat is supplied, this process is also known as thermal
20 stripping. However, the decomposition preferably is effected by stripping with the aid of a stripping gas in countercurrent to the urea synthesis solution, with addition of heat. Ammonia, carbon dioxide, and an inert gas, alone
25 or in any combination, can be used as the stripping gas.

The stripping operation can be carried out at a pressure that is the same as the synthesis pressure or at a slightly higher or lower
30 pressure. Preferably substantially the same pressures are used in the reactor and in the stripping zone, since this makes it possible to return the gases formed in the stripping zone to the reactor in a non-complex manner.

35 In another embodiment of the present invention the gases from the stripper are used to convert water into steam or to heat a process

liquid in a first heat exchanger disposed outside the reactor, after which the partially condensed gases are sent to the reactor according to the invention. In this manner, at most about 70% and preferably at most about 50% of the energy to be discharged from condensation is discharged in this first heat exchanger; correspondingly, at least about 30%, and preferably more than about 50%, of the heat of condensation is discharged in the heat exchanger of the reactor. This embodiment is advantageous when, for example, a process stream needs to be heated (for example a urea solution to be concentrated) and the generation of steam as a by-product is desirable.

A particularly advantageous embodiment of the present invention involves preparing the urea in a reactor with a condensation zone and a heat exchanger, in which the gases from the stripper are returned directly to the condensation zone. In this case all of the energy to be discharged is discharged in and transferred to the heat exchanger of the reactor. This is particularly preferable because of the simplicity, and hence inexpensive nature of the design.

Preferably carbon dioxide (CO_2) is used as the stripping gas so that the stripping gas is hence used as the CO_2 -containing gas that is fed to all the reactor's compartments. Besides CO_2 , this stripping gas contains NH_3 and inert gas. In addition to all the extra liquid ammonia to be supplied, preferably more than about 60% of this stripping gas, in particular more than about 70%, is fed to the condenser zone of the reactor.

The present invention is illustrated by the following figures and the examples, without being limited thereto.

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As shown in FIG. 1, A represents a reactor with a condensation zone and a heat exchanger, B a stripping zone, C a steam reservoir, and D, E, F and G pumps or compressors.

5 Pump E supplies compressed liquid ammonia to the reactor's condensation zone via conduit (1) and via a pipe (4) provided with openings. A carbamate solution that has been obtained elsewhere in the process, in particular by washing
10 off-gases with an aqueous solution obtained in the evaporation of the urea solution, is introduced into the reactor (A) via conduits (2) and (3) by means of pump D. A gaseous mixture containing ammonia and carbon dioxide is introduced into the
15 liquid via a pipe (5) provided with openings. This gaseous mixture - supplied via pipe (15) - is obtained by subjecting the urea synthesis solution formed in the reactor (A) to a stripping treatment in the stripping zone (B), with the addition of
20 heat and in countercurrent contact to a stripping gas, for example carbon dioxide, supplied via pipe (13). The pressures in the reactor (A) and the-stripping zone (B) are the same in the embodiment shown, for example about 140 bar. The
25 pressures in these zones may, however, differ from one another. The dimensions of the reactor (A) are selected so that the residence time of the reaction mixture in this reactor is sufficiently long to ensure that at least about 85% of the
30 theoretically possible amount of urea is formed in the reactor. The reactor (A) is provided with partitions (16) (or (23) in FIGS. 2 and 3) which divide the reactor into compartments. The last partition (i.e., located at the far left of the
35 reactor depicted in FIG. 1) has an aperture at the upper portion thereof only to enable the level of the condensate in the last compartment to be

controlled. At startup the penultimate and last compartments must then be (temporarily) connected to one another via a bypass pipeline (17), so that a urea synthesis solution can already be
5 discharged when the reactor is half full.

The heat that is released in the reactor (A) is discharged with the aid of water, which is supplied via conduit (6), which is passed through the heat exchanger (8) installed in the reactor
10 (A) by means of pump G via conduit (7). The water is converted into low-pressure steam in that process. The steam formed is sent to steam reservoir C via conduit (9) and is discharged therefrom via conduit (10) to a low-pressure
15 steam-consuming installation (not shown) which can be, for example, the recycling and/or evaporation section. Instead of discharging the heat as a steam formation, it is also possible to pass a process stream that needs to be heated, for
20 example an aqueous urea solution that is to be concentrated, such as stream (12), through the cooling elements.

The inert gases which contain ammonia and CO_2 are discharged from the reactor (A) via outlet
25 (14). NH_3 and CO_2 are removed from these gases in a known manner. The urea synthesis solution passes from the reactor (A) via conduit (11) to the stripping zone (B). The stripped urea synthesis solution is discharged via stream (12) and can be
30 further processed to an aqueous urea solution and concentrated in a known manner, whereupon the concentrated solution is optionally converted into solid urea.

In FIGS. 2 and 3, which represent
35 cross-sections of the reactor A, (21) represents the reactor wall. For practical reasons there can be an opening (22) defined between the partitions

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(23) which divide the reactor into compartments and the wall (21). The gas from the stripper is introduced into the reactor A through the distributing device (24). For practical reasons there are manholes (25) and (25') in each partition. When the reactor is in operation, the manholes may be open or closed, depending on the desired throughput of the urea synthesis solution. The manholes in the partitions near the heat exchanger (29) are preferably closed, while each of the remaining partitions preferably has only one manhole open, with the open manholes being alternated a staggered fashion so that a zigzag liquid stream is obtained. The top side of a partition (23) defines an opening (26) as a gas discharge area. The partitions may be provided with vertical baffle partitions (27), which are however not essential. In Figure 2 ammonia is introduced into the reactor by means of the distributing device (28) at the level of the condenser/heat exchanger and the heat exchanger is represented as (29).

A method for the preparation of urea is disclosed in NL 1,000,416.

The following non-limiting examples serve to explain embodiments of the present invention in more detail.

Example

A plant with a high-pressure synthesis unit as shown in Fig. 1 was used to produce 70 tons of urea per hour. The pressure was 144 bar; under these conditions the degree of conversion in the reactor was 95% relative to the theoretical equilibrium. In the heat exchanger 78.7 tons of steam of 4.5 bar (148°C) was produced per hour

from 78.7 tons of condensate of 175°C; this corresponds to some 600 kWh per ton of urea.

The streams through the pipelines were as follows.

5	Compo- nent	Amounts in tons / hour						
		1	2	11	12	13	14	15
	Urea			75.2	70.0			
	NH ₃	39.7	20.8	62.9	8.7		12.1	57.1
10	CO ₂		17.1	35.8	11.7	51.3	5.4	79.2
	H ₂ O		9.7	36.5	30.2		0.6	4.8
	N ₂					1.3	1.3	1.3
	O ₂					0.2	0.2	0.2
	TOTAL	39.7	47.6	210.3	120.5	52.9	19.7	142.7
15	Temp. (°C)	20		186	173	120	176	187

The stripped urea synthesis solution obtained from pipeline (12) was further processed into solid urea in a manner known to those skilled in the art.

Although the present invention has been described in detail with reference to its presently preferred embodiments, it will be understood by those of ordinary skill in the art that various modifications and improvements to the present invention are believed to be apparent to one skilled in the art. Accordingly, no limitation upon the invention is intended, except as set forth in the appended claims.

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WHAT IS CLAIMED IS:

1. A method for the preparation of urea, the urea being formed along with ammonium carbamate from carbon dioxide and ammonia in a synthesis zone at a pressure of about 125 bar to about 350 bar, said method comprising the steps of:

providing a reactor comprising the synthesis zone, a horizontally arranged condensation zone, and a heat exchanger;

supplying ammonia and carbon dioxide to the reactor so that the ammonia and the carbon dioxide are substantially absorbed into a urea synthesis solution;

maintaining the urea synthesis solution in the reactor for a residence time selected so that at least about 85% of a theoretically obtainable amount of urea is prepared; and

discharging a substantial portion of heat formed from condensation in the condensation zone to a fluid passing through the heat exchanger.

2. A method according to Claim 1, wherein the residence time of the urea synthesis solution is selected so that at least about 90% of the theoretically obtainable amount of urea is prepared.

3. A method according to Claim 1, wherein the reactor is arranged horizontally.

4. A method according to Claim 3, wherein the reactor is provided with means that ensure that the liquid flows through the reactor substantially in plug flow.

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5. A method according to 2, wherein the reactor has a pipe shape, with a diameter of between about 1 m and about 5 m and a length of between about 5 m and about 40 m.

6. A method according to Claim 5, wherein the reactor is provided with a structured packing or partitions which divide the reactor into compartments.

7. A method according to Claim 6, further comprising a step of introducing gas into the compartments to generate turbulence.

8. A method according to Claim 1, further comprising a step of supplying liquid ammonia to a portion of the reactor containing both the condensation zone and the heat exchanger.

9. A method according to Claim 1, further comprising a step of supplying at least about 60% of the carbon dioxide to a portion of the reactor containing both the condensation zone and the heat exchanger.

10. A method for the preparation of urea, the urea being formed along with ammonium carbamate from carbon dioxide and ammonia in a synthesis zone at a pressure of about 125 bar to about 350 bar, said method comprising the steps of:

providing a reactor comprising the synthesis zone, a horizontally arranged condensation zone, and a heat exchanger;

supplying ammonia and carbon dioxide to the reactor so that the ammonia and the carbon dioxide

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are substantially absorbed into a urea synthesis solution;

maintaining the urea synthesis solution in the reactor for a residence time selected so that at least about 85% of a theoretically obtainable amount of urea is prepared;

removing at least a portion of the carbamate from the reactor and decomposing the carbamate in a stripping zone to form carbamate decomposition products;

passing the carbamate decomposition products to the reactor for condensation in the condensation zone thereof;

discharging a substantial portion of heat formed from the condensation to a fluid passing through the heat exchanger.

11. A method according to Claim 10, wherein a CO₂-containing gas is obtained in the stripping zone and returned to the reactor.

12. A method according to Claim 11, wherein heat that is discharged in the heat exchanger of the reactor amounts to more than about 125 kWh per ton of the urea produced.

13. A method according to Claim 10, wherein the residence time of the urea synthesis solution is selected so that at least about 90% of the theoretically obtainable amount of urea is prepared.

14. A method according to Claim 10, wherein the reactor is arranged horizontally.

15. A method according to Claim 14, wherein the reactor is provided with means that ensure

that the liquid flows through the reactor substantially in plug flow.

16. A method according to 13, wherein the reactor has a pipe shape, with a diameter of between about 1 m and about 5 m and a length of between about 5 m and about 40 m.

17. A method according to Claim 16, wherein the reactor is provided with a structured packing or partitions which divide the reactor into compartments.

18. A method according to Claim 17, further comprising a step of introducing gas into the compartments to generate turbulence.

19. A method according to Claim 10, further comprising a step of supplying liquid ammonia to a portion of the reactor containing both the condensation zone and the heat exchanger.

20. A method according to Claim 10, further comprising a step of supplying at least about 60% of the carbon dioxide to a portion of the reactor containing both the condensation zone and the heat exchanger.

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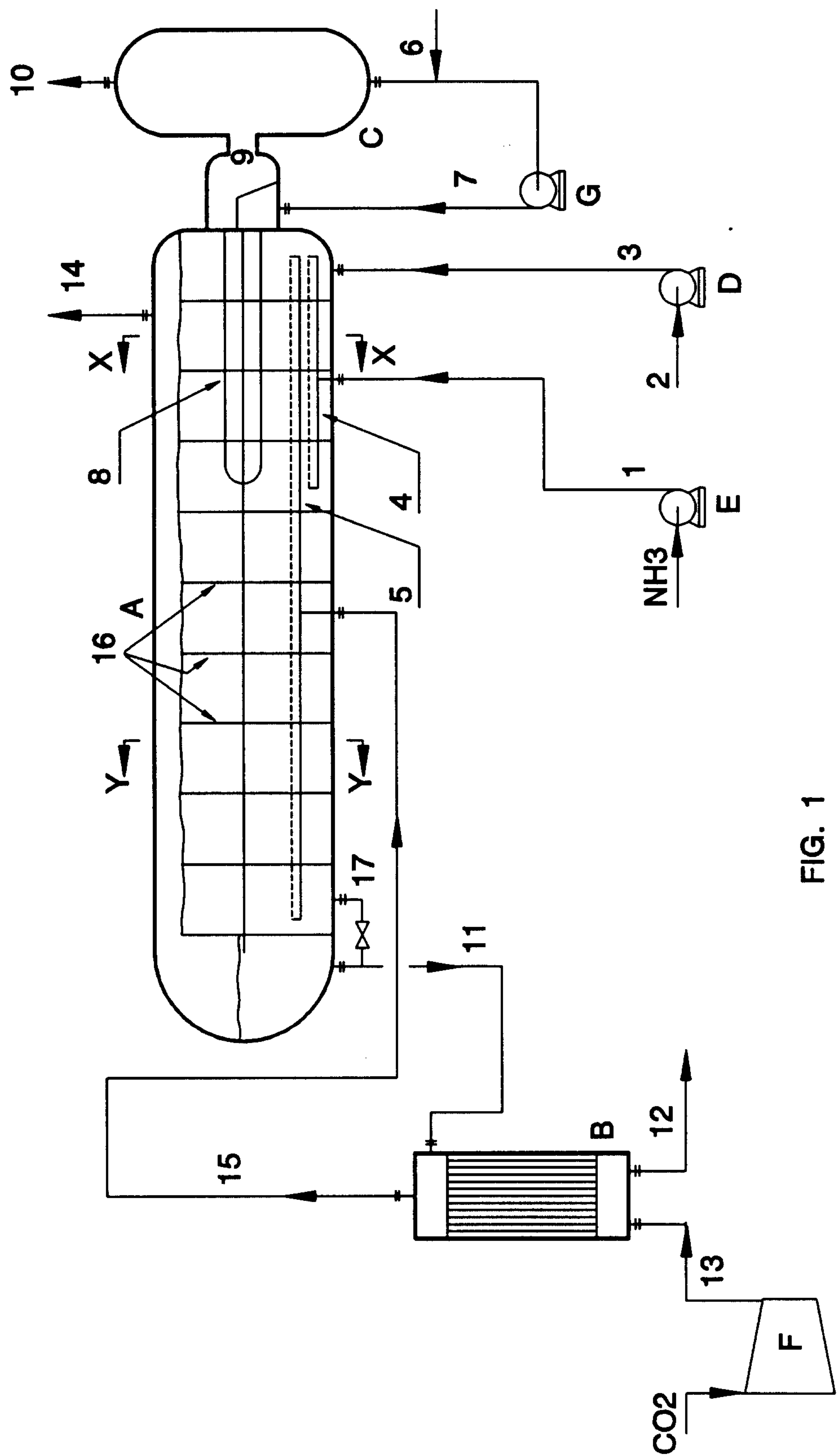
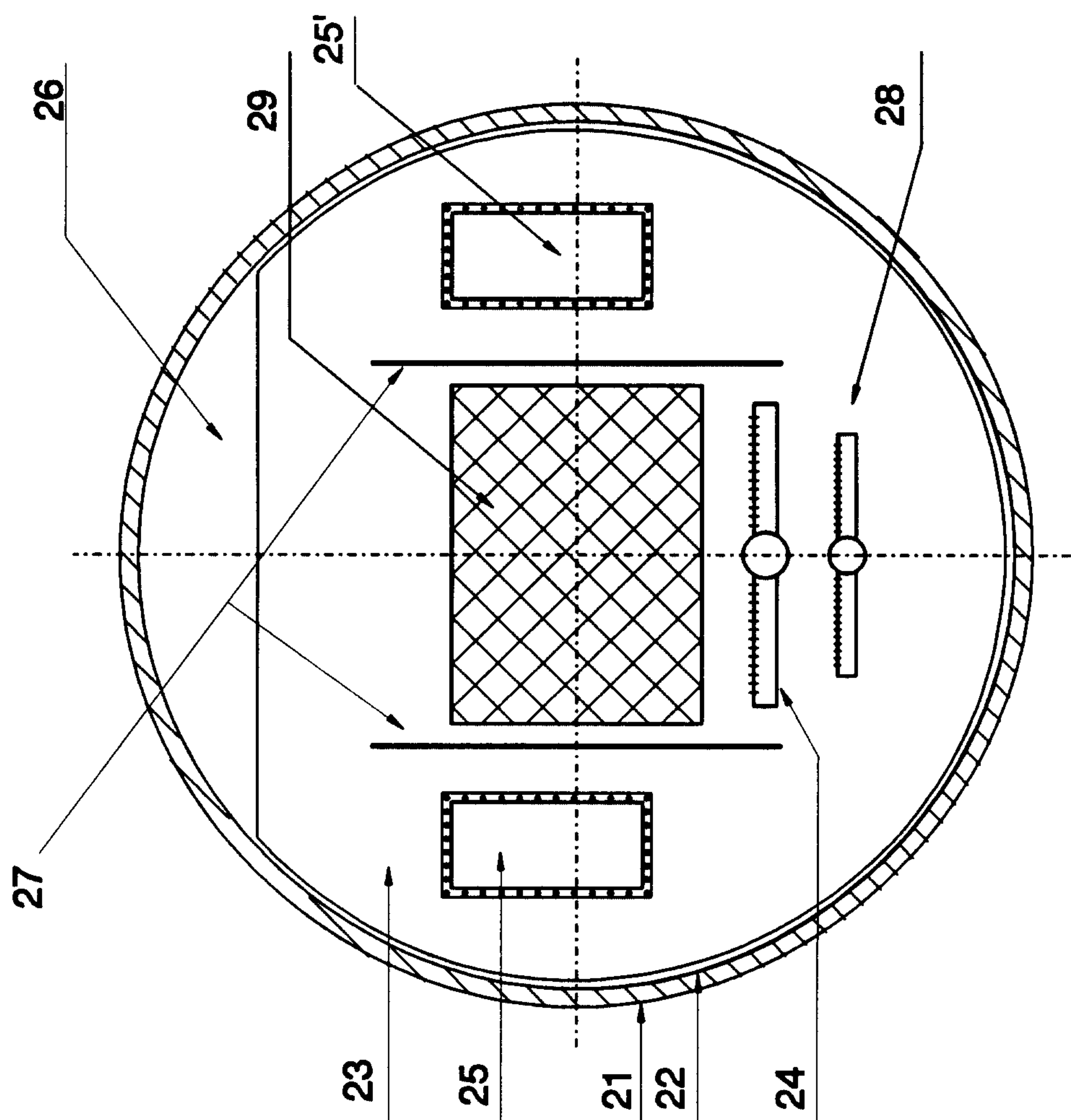


FIG. 1

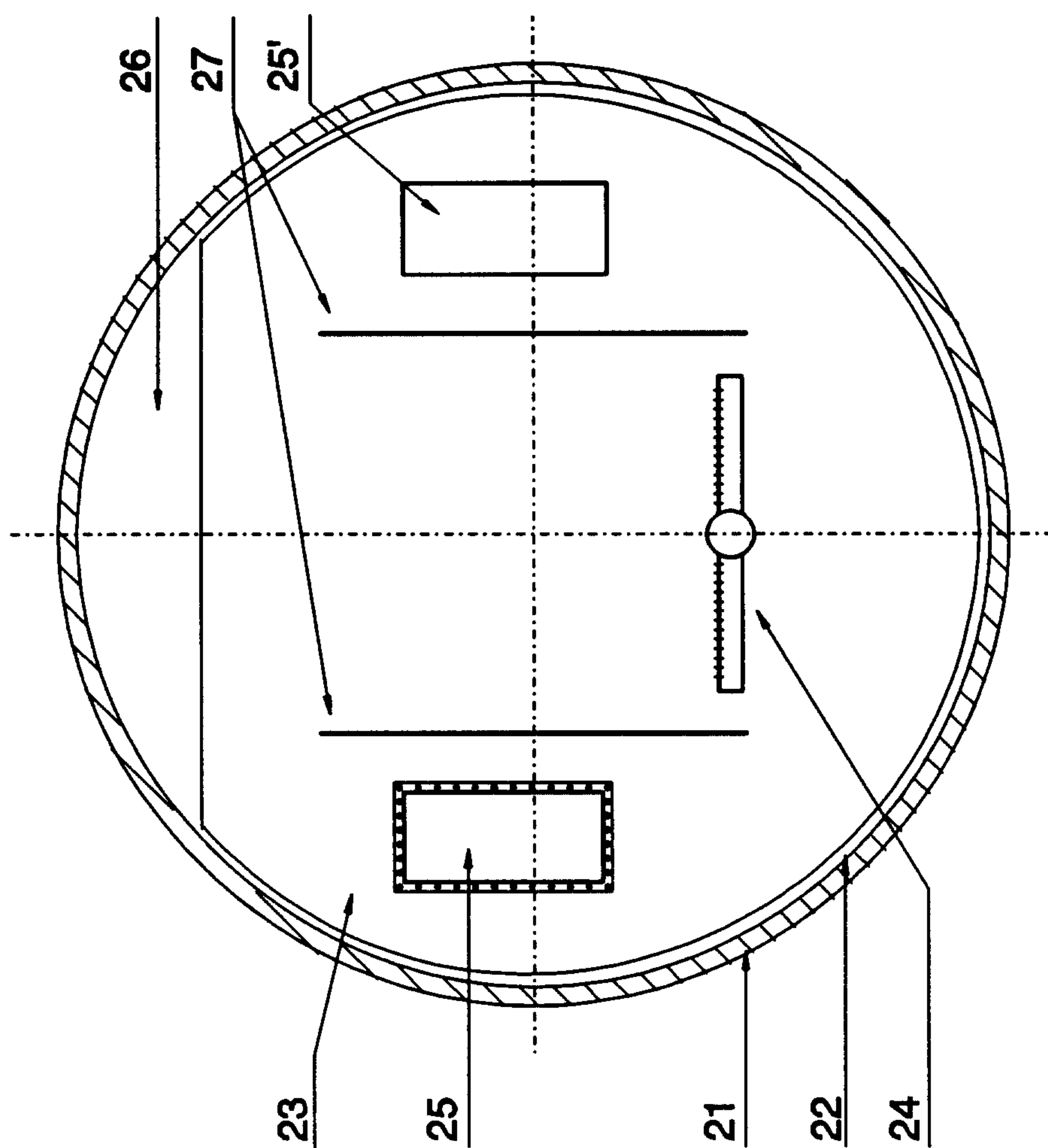
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X-X
FIG. 2

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Y-Y
FIG. 3

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